

### **REMARKS**

The Official Action dated July 7, 2008 has been carefully considered. Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present Amendment, the specification is amended to include the §371 information from the Inventors' Declaration and the Official Filing Receipt. Claim 1 is amended to recite that the sulfur source comprises an alkali metal hydrosulfide alone or a mixture of 90 to 99.5 mol% of an alkali metal hydrosulfide and 0.5 to 10 mol% of an alkali metal sulfide, as set forth in the present specification at page 12, lines 20-26. Additionally, claim 1 is amended to clarify that in the polymerization step, the pH of the mixture for polymerization reaction is measured by collecting the reaction mixture, diluting it to 1/10 with ion-exchanged water and measuring the pH of the diluted solution in accordance with the teachings from the present specification at page 28, line 25 to page 29, line 1. Claims 3, 6 and 8 are amended for matters of form. It is therefore believed that no new matter is introduced by these amendments, whereby entry of the Amendment is believed to be in order and is respectfully requested.

In the Official Action, the Examiner rejected claims 1-12 under 35 U.S.C. §102(b) as anticipated by or, alternatively, under 35 U.S.C. §103(a) as obvious over, Miyahara et al (US Patent No. 5,840,830). The Examiner asserted that Miyahara et al disclose a process for producing a poly(arylene sulfide) (PAS) comprising a dehydration step of heating and dehydrating a mixture containing an organic amide solvent, an alkali metal sulfide and water, and a subsequent polymerization step of reacting the alkali metal sulfide and a dihalo-aromatic compound in a polymerization reaction. The Examiner noted that Miyahara et al disclose that an alkali metal hydrosulfide and an alkali metal hydroxide may be added in almost equimolar

amounts to provide the alkali metal sulfide in situ. The Examiner admitted that Miyahara et al do not disclose the step of adding the alkali metal hydroxide to the mixture for polymerization continuously or in portions to control the pH within a range of 7 to 12.5 but the Examiner asserted this would be inherent in Miyahara et al's teachings since they disclose a similar step.

This rejection is traversed and reconsideration is respectfully requested. Miyahara et al do not inherently disclose a production process as presently claimed and there is no apparent reason for one of ordinary skill in the art to modify the teachings of Miyahara et al to arrive at the claimed process.

More particularly, as defined by claim 1, the present invention is directed to a process for producing a poly(arylene sulfide) (PAS) by polymerizing a sulfur source and a dihalo-aromatic compound in the presence of an alkali metal hydroxide in an organic amide solvent. The process comprises a dehydration step and a polymerization step. The dehydration step comprises charging, into a reaction vessel, the organic amide solvent and the sulfur source, and a part of an overall charged amount of the alkali metal hydroxide as needed, and heating a mixture containing these components to discharge at least a part of a distillate containing water from the interior of the system containing the mixture to the exterior of the system. The sulfur source comprises (i) an alkali metal hydrosulfide alone, or (2) a mixture of 90 to 99.5 mol% of an alkali metal hydrosulfide and 0.5 to 10 mol% of an alkali metal sulfide.

The polymerization step comprises mixing the mixture remaining within the system after the dehydration step with a dihalo-aromatic compound, heating a mixture for polymerization reaction containing these components to subject the sulfur source ("available sulfur source") and the dihalo-aromatic compound to a polymerization reaction, and adding the alkali metal hydroxide to the mixture for polymerization reaction continuously or in portions to control the

pH of the mixture for polymerization reaction within a range of from 7 to 12.5 from the beginning to the end of the polymerization reaction. The pH of the mixture for polymerization reaction is measured by collecting the reaction mixture, diluting it to 1/10 with ion-exchanged water and measuring the pH of the diluted solution.

Miyahara et al disclose a process for producing a poly(arylene sulfide) which comprises a dehydration step of heating and dehydrating a mixture containing an organic solvent (a), an alkali metal sulfide and water to control a water content in the mixture, and a subsequent polymerization step of subjecting the alkali metal sulfide and a dihalo-aromatic compound to a polymerization reaction in the organic amide solvent (a). Hydrogen sulfide vaporized off during the dehydration step is absorbed in another organic amide solvent (b) outside the system in which the dehydration step is carried out, thereby recovering it, and the hydrogen sulfide thus recovered is reused in the polymerization reaction as a raw material for the alkali metal sulfide.

The production process according to the present invention is neither anticipated by nor rendered obvious over, and is significantly different from, the production process disclosed by Miyahara et al. Noting that NaHS, Na<sub>2</sub>S and NaOH are respectively representative of the alkali metal hydrosulfide, alkali metal sulfide and alkali metal hydroxide, there are two important distinctions between the present process and that of Miyahara et al. First, the present process employs an alkali metal hydrosulfide (NaHS) alone or a mixture of 90 to 99.5 mol% of an alkali metal hydrosulfide (NaHS) and 0.5 to 10 mol% of an alkali metal sulfide (Na<sub>2</sub>S) as a sulfur source charged in the production process, whereas only an alkali metal sulfide (Na<sub>2</sub>S) is used as a sulfur source charged in the production process as exemplified by Miyahara et al. While Miyahara et al generally disclose that an alkali metal hydrosulfide may be used to prepare the alkali metal sulfide in situ, Miyahara et al do not exemplify such a process and therefore do not

describe sufficient details of such a process to support a rejection based on inherency. Second, in the present process polymerization step, an available sulfur source and a dihalo-aromatic compound are subjected to a polymerization reaction, and an alkali metal hydroxide (NaOH) is added to the mixture for the polymerization reaction continuously or in portions to control the pH of the mixture for the polymerization reaction within a range of from 7 to 12.5 from the beginning to the end of the polymerization reaction, whereas Miyahara et al neither inherently disclose nor suggest that the alkali metal hydroxide (NaOH) is added to the mixture for polymerization reaction continuously or in portions to control the pH of the mixture for polymerization reaction within a range of from 7 to 12.5 from the beginning to the end of the polymerization reaction. These differences and their nonobvious effects on the respective processes are discussed in detail below.

As described in the “BACKGROUND ART” section of the present specification, a poly(arylene sulfide) (PAS) production process using an alkali metal hydrosulfide (NaHS) as a sulfur source encounters unique problems as compared with a conventional PAS production process which uses an alkali metal sulfide ( $\text{Na}_2\text{S}$ ). For example, in a typical PAS production process, a sulfur source is reacted with a dihalo-aromatic compound in an organic amide solvent such as N-methyl-2-pyrrolidone (NMP). However, when an alkali metal hydrosulfide (NaHS) and an alkali metal hydroxide (NaOH) are used in combination as materials for the sulfur source, the organic amide solvent is easily decomposed because the alkali metal hydroxide (NaOH) is used at a high concentration in the polymerization reaction. In addition, the resulting decomposition products cause undesirable side reactions.

For example, when NMP is used as an organic amide solvent, it is decomposed by an alkali metal hydroxide (NaOH) present in a high concentration and a salt of a

methylaminoalkylcarboxylic acid is formed and causes undesirable side reactions whereby low-boiling impurities are contained in the resulting PAS. As a result, when PAS obtained by such a process is subjected to melt processing, gasses are undesirably generated, thereby clogging exhaust lines in melt-processing apparatus and/or facilitating production of voids in molded and formed products. When voids are produced in a molded product, the physical properties and surface properties thereof are deteriorated.

The above-described tendencies may be generally observed when an alkali metal hydrosulfide (NaHS) is used as a sulfur source and an alkali metal hydroxide (NaOH) is used at a higher molar ratio than the alkali metal hydrosulfide for the purpose of providing a PAS having a high melt viscosity. On the other hand, when the alkali metal hydroxide (NaOH) is used at a lower molar ratio than the alkali metal hydrosulfide (NaHS), the polymerization reaction system becomes unstable and a decomposition reaction easily occurs. Accordingly, in conventional production processes, a high molar ratio of the alkali metal hydroxide (NaOH) to the alkali metal hydrosulfide (NaHS) is used to conduct the polymerization reaction, but side reactions as described typically occur and it is therefore difficult to produce a high yield of high-purity PAS.

Miyahara et al describe that

“The alkali metal sulfide may also be prepared in situ in an organic amide solvent from an alkali metal hydrosulfide and an alkali metal hydroxide. It may also be possible to use a small amount of an alkali metal hydroxide in combination with the alkali metal sulfides to react it with alkali metal hydrosulfides and alkali metal thiosulfates, which may sometimes exist in a trace amount in the alkali metal sulfides, thereby removing these trace components or converting them into the alkali metal sulfides.” (column 4, line 61 to column 5, line 3),

and

“When an alkali metal hydrosulfide is used as a sulfur source, an alkali metal hydroxide is added in an amount almost equimolar thereto, thereby reacting both components in situ in the organic amide solvent to convert into its corresponding alkali metal sulfide. In this reaction, water is formed as a by-product. In the dehydration step, water composed of

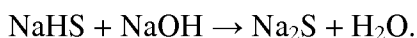
hydrate (water of crystallization), water as a medium for the aqueous mixture and water formed as the by-product is removed until the water content in the polymerization reaction system comes within a range of necessary amounts.” (column 6, lines 49-59).

However, the Miyahara et al Examples only employ an alkali metal sulfide ( $\text{Na}_2\text{S}$ ) as a sulfur source, and none of the Miyahara et al Examples employ an alkali metal hydrosulfide ( $\text{NaHS}$ ) as a sulfur source. Miyahara et al do not specifically disclose the use of an alkali metal hydrosulfide ( $\text{NaHS}$ ) alone or a mixture of 90 to 99.5 mol% of an alkali metal hydrosulfide ( $\text{NaHS}$ ) and 0.5 to 10 mol% of an alkali metal sulfide ( $\text{Na}_2\text{S}$ ) as a sulfur source as recited in claim 1. Quite naturally, then, Miyahara et al neither recognize the various problems as described above which result from the combined use of the sulfur source comprising the alkali metal hydrosulfide ( $\text{NaHS}$ ) as a main component and the alkali metal hydroxide ( $\text{NaOH}$ ), nor propose means for solving these various problems and particularly do not teach or suggest a pH control step as required by claim 1 to avoid these problems. While Applicants recognize that a reference is not limited to exemplary teachings, any rejection based on inherency must establish that the claim limitations are necessarily present in the teachings of the prior art, *In re Oelrich*, 666 F.2d 578 (CCPA 1981), MPEP §2112. The present claim limitations are not necessarily present in Miyahara et al.

Importantly, the present specification demonstrates that when the proportion of alkali metal hydroxide ( $\text{NaOH}$ ) employed in the Examples of Miyahara et al is used, the pH of the mixture for polymerization reaction is not controlled within the range of from 7 to 12.5 from the beginning to the end of the polymerization reaction as required by claim 1. That is, Example 1a of Miyahara et al describes a PAS production process using sodium sulfide ( $\text{Na}_2\text{S}$ ) as a sulfur source, wherein i) dehydration was conducted after 30 g of 97%  $\text{NaOH}$  was added in a dehydration step, and then ii) 7.9 g of 97%  $\text{NaOH}$  was added in a polymerization step to control

the total amount of NaOH (including NaOH formed by the vaporization of H<sub>2</sub>S) in the vessel to 5.00 mol % of the available Na<sub>2</sub>S. In other Examples of Miyahara et al, NaOH/Na<sub>2</sub>S (available sulfur source) was controlled to 5.00 mol%, 7.50 mol% and 7.60 mol%, respectively (see Table 1). Miyahara et al do not otherwise describe the range of NaOH/Na<sub>2</sub>S (available sulfur source), and the above numerical values are only shown in the Examples.

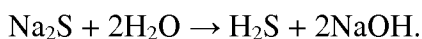
However, the pH of the mixture for the polymerization reaction at the beginning of the polymerization reaction exceeds the claim 1 upper limit of 12.5 when a proportion of NaOH/Na<sub>2</sub>S of 5.00 mol% is employed. In this regard, the Examiner's attention is directed to Comparative Example 1 beginning at page 46 of the present specification, which shows a PAS production process using, as a sulfur source, NaSH (21.8 mol) and Na<sub>2</sub>S (0.50 mol). In the dehydration step of this Comparative Example 1, 1,170 g of an aqueous solution of NaOH having a concentration of 74.7% by weight is added. The amount of NaOH in this aqueous solution is 21.8 mol. Accordingly, NaHS in an amount of 21.8 mol is reacted with the 21.8 mol of NaOH to form 21.8 mol of Na<sub>2</sub>S in accordance with the reaction scheme:



Thus, the entire amount of NaOH added in the dehydration step is consumed in the reaction with NaHS. Since 0.50 mol of Na<sub>2</sub>S is contained in the sulfur source charged, the total amount of Na<sub>2</sub>S is as follows:

$$\text{Total Na}_2\text{S} = 21.8 + 0.50 = 22.3 \text{ mol}.$$

In the dehydration step of Comparative Example 1, 0.31 mol of hydrogen sulfide (H<sub>2</sub>S) is vaporized off in accordance with the reaction scheme:



Accordingly, an equimolar amount (0.31 mol) of  $\text{Na}_2\text{S}$  is lost by vaporization of 0.31 mol of hydrogen sulfide ( $\text{H}_2\text{S}$ ), while a double molar amount ( $0.31 \times 2 = 0.62$  mol) of  $\text{NaOH}$  is formed, thereby obtaining the following respective values:

$$\text{Available S} = 22.3 - 0.31 = 22.0 \text{ mol}$$

$$\text{generated NaOH} = 0.62 \text{ mol, and}$$

$\sum \text{OH} = 22.6$  mol (0.50 mol of charged  $\text{Na}_2\text{S}$  + 21.8 mol of added  $\text{NaOH}$  + 0.31 mol of volatilized  $\text{H}_2\text{S}$ ).

$\sum \text{OH}$  is a value calculated out on the basis of the definition described in claim 6 of the present application.

The polymerization step of Comparative Example 1 similarly employ an  $\text{NaOH}/\text{Na}_2\text{S}$  molar ratio of 5.00 mol%. That is, 23 g of  $\text{NaOH}$  having a concentration of 97% by weight was added in such a manner that the value of  $\sum \text{OH}/\text{available S}$  is 1.05. The molar amount of  $\text{NaOH}$  added is 0.56 mol. The molar ratio of  $\text{NaOH}$  to available  $\text{Na}_2\text{S}$  calculated for the components in Comparative Example 1 of the present specification in accordance with the method disclosed in Miyahara et al, is 5%, since the available  $\text{S} = 22.0$  mol, and the amount of  $\text{NaOH}$  in the vessel is 0.62 mol (dehydration step) + 0.56 mol (polymerization step) = 1.18 mol.

$$\text{NaOH}/\text{Na}_2\text{S} (\text{available sulfur source}) = 1.18/22.0 = 0.05 (\text{NaOH} = 5 \text{ mol } \%).$$

As noted above, this is the lowest ratio employed by Miyahara et al in the Miyahara et al Examples. However, Comparative Example 1 of the present specification further shows that when the proportion of  $\text{NaOH}$  to the available  $\text{S}$  was 5 mol %, the pH of the mixture for polymerization reaction was 13.2 (see page 47). It is therefore apparent that the pH of the Miyahara et al mixture for polymerization reaction exceeds 12.5 when the reaction conditions specifically disclosed in Miyahara et al are adopted. Thus, the step of adding the alkali metal



hydroxide to the mixture for polymerization continuously or in portions to control the pH within a range of 7 to 12.5 is not inherent in the reaction conditions of Miyahara et al, even if an alkali metal hydrosulfide is employed. Miyahara et al therefore do not inherently anticipate or render obvious the claimed production process.

The present specification further shows in Comparative Example 3-1 that a high pH above 12.5 in the reaction mixture provides PAS with an undesirable high nitrogen content and low melt viscosity. According to the results shown in Table 1 at page 51 of the present specification (Comparative Example 1) and Table 2 at page 58 (Comparative Example 3-1), it is evident that when a sulfur source comprising an alkali metal hydrosulfide (NaHS) as a main component is used, and the pH of the mixture for polymerization reaction is controlled to 13.2, thereby exceeding the upper limit of claim 1 of 12.5, to initiate a polymerization reaction, the polymerization reactivity and the physical properties and properties of the resulting PAS are lowered.

According to the production process of the present invention, however, PAS having markedly reduced low-boiling impurities content can be produced with high production efficiency and without deterioration of profitability due to loss of raw materials, or the like. Further, according to the production process of the present invention, PAS having a high molecular weight as evidenced by high melt viscosity can be produced. The resulting PAS also has a low nitrogen content, little variation in melt viscosity due to differences in washing liquid and resin pH, and a stable, desired melt viscosity.

In determining patentability under 35 U.S.C. §103, it is necessary to determine whether there was an apparent reason to combine the known elements in the fashion of the claim at issue, *KSR International Co. v. Teleflex, Inc.*, 127 S.Ct. 1727, 1740-41 (2007). Miyahara et al neither

teach nor suggest that in order to achieve the above improvements, a sulfur source comprising an alkali metal hydrosulfide as a main component is used and the pH of the mixture for polymerization reaction is controlled within a range of from 7 to 12.5 from the beginning to the end of the polymerization reaction as required by claim 1. Accordingly, Miyahara et al provide no apparent reason to one of ordinary skill in the art to modify the Miyahara et al teachings to result in the production processes defined by claims 1 and 3-12. Thus, the claimed production processes are not rendered obvious over Miyahara et al, whereby the rejections under 35 U.S.C. 102(b) and 103(a) have been overcome. Reconsideration is respectfully requested.

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested. Please charge any fee required with this response to Deposit Account No. 503915.

Respectfully submitted,

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